



Gel polymer electrolyte for lithium-ion batteries comprising cyclic carbonate moieties



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HIGHLIGHTS

- Synthesis of a new polymer matrix comprising cyclic carbonate moieties.
- Polymer matrix is able to retain huge amounts of liquid electrolyte.
- Gel polymer electrolyte displays high conductivity and good mechanical stability.
- Gel polymer electrolyte shows good performance in NCM/graphite full cells.

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ABSTRACT

A polymer system based on oligo (ethylene glycol) methyl ether methacrylate (OEGMA) and cyclic carbonate methacrylate (CCMA) was chosen as matrix to realize high-performance gel polymer electrolytes due to the fact that both monomers are able to interact with the liquid electrolyte, thus, retaining it inside the matrix. Additionally, OEGMA enables high flexibility, while CCMA provides mechanical stability. The polymer displays a high thermal stability up to 200 °C and a glass transition temperature below room temperature (5 °C) allowing an easy handling of the obtained films. By immobilizing the liquid electrolyte 1 M LiPF₆ in EC:DMC 1:1 w:w in the polymer host a gel polymer electrolyte with a high conductivity of 2.3 mS cm⁻¹ at 25 °C and a stable cycling behavior with high capacities and efficiencies in Li(Ni_{1/3}Co_{1/3}Mn_{1/3})O₂ (NCM)/graphite full cells is obtained. The investigated gel polymer electrolyte is identified as promising electrolyte for lithium-ion batteries, because it combines good electrochemical properties comparable to that of liquid electrolytes with the safety advantage that no leakage of the flammable electrolyte solvents can occur.

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1. Introduction

Lithium-ion batteries – characterized by their high discharge voltage of about 3–4 V and their unbeatable specific energy of 150 Wh kg⁻¹ – are already the favored battery system for the portable market [1,2]. Moreover they have succeeded in entering the sustainable vehicle market [3]. Despite the unique features they offer, lithium-ion batteries have not yet reached the technological level needed for a large-scale commercialization for electric and hybrid-electric vehicles. Recent studies focus on the improvement of energy density and safety, which comes along with the development of materials for the various battery components [4]. While the electrode material stands out due to high capacity and

durability, the electrolyte needs to have a high ionic conductivity at a broad temperature range of use [5]. Furthermore, properties like a wide electrochemical stability window and environmental friendliness are indispensable for future developments.

Commercial lithium-ion battery electrolytes usually contain a lithium salt, typically LiPF₆, which is dissolved in mixtures of cyclic and linear carbonates as solvents [6,7]. Despite the positive features like high conductivity and stability in a wide temperature range, these electrolytes show some issues, especially concerning large-scale applications for electromobility and stationary energy storage.

Using liquid electrolytes leads to safety hazards such as fire and explosion, which can be caused by leakage of liquid electrolyte comprising highly flammable linear carbonates as solvent [8]. Solid polymer electrolytes, which consist of a polymer (e.g. polyethylene oxide) in which a lithium salt is dissolved, would be an ideal candidate to make batteries safer, but this advantage goes along with a considerably reduced conductivity [9]. To combine the

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positive properties of liquid and solid polymer electrolytes, gel polymer electrolytes have been introduced. They consist of a liquid electrolyte, which is immobilized in a polymer matrix. Therefore, leakage can be avoided [10]. Recently, our group reported on the preparation of statistical co-polymer membranes via UV-induced free radical polymerization comprising oligo (ethylene glycol) methyl ether methacrylate (OEGMA) and benzyl methacrylate (BnMA) as monomers. Due to its ethylene glycol side chains, OEGMA is able to interact with the liquid electrolyte [11], whereas BnMA was used to enhance the mechanical stability of the gel polymer electrolyte. Gel polymer electrolytes based on this co-polymer showed promising performances in lithium-ion batteries as well as in high power devices [12,13].

In this paper a polymer host based on OEGMA and cyclic carbonate methacrylate (CCMA) is synthesized and gelled using the state-of-the-art liquid electrolyte 1 M LiPF₆ in ethylene carbonate (EC):dimethyl carbonate (DMC) 1:1. CCMA was chosen as monomer as it displays a similar structure like the solvents of the liquid electrolyte. Therefore, it is assumed that CCMA is able to interact with the liquid electrolyte, which would be beneficial for the liquid electrolyte uptake capacity as well as for the ability to retain the liquid electrolyte inside the polymer matrix. Apart from thermal and electrochemical investigations of the gel polymer electrolyte, the applicability in lithium-ion batteries is evaluated.

2. Experimental

2.1. Chemicals

Glycerol 1,2-carbonate ($\geq 90\%$) was obtained from TCI, triethylamine ($\geq 99.6\%$), ethyl acetate (EtOAc, 99%) and pentane ($\geq 99\%$) from VWR International and methacryloyl chloride ($\geq 95\%$) and benzophenone (BNP, $\geq 99\%$) from Acros Organics. Butylated hydroxytoluene (BHT, $\geq 99\%$) was purchased from Fluka and oligo (ethylene glycol) methyl ether methacrylate ($M_n = 300 \text{ g mol}^{-1}$) from Sigma–Aldrich.

2.2. Monomer synthesis

Cyclic carbonate methacrylate (CCMA) was synthesized according to a slightly modified method described by Kilambi et al. (see Scheme 1) [14]. Glycerol 1,2-carbonate (3.54 g, 0.030 mol, 1.00 eq.), triethylamine (4.44 mL, 0.032 mol, 1.07 eq.) and EtOAc (30 mL) were introduced into a three-neck flask, which was constantly purged with argon. To maintain the reaction temperature at a low level, the flask was immersed in an ice-water bath. The dropping funnel was charged with methacryloyl chloride (2.71 mL, 0.028 mol, 0.93 eq.) in ethyl acetate (5 mL) and the solution was added dropwise within two hours. Afterward, the reaction mixture was stirred over night at RT. For purification the reaction mixture was filtered, washed several times with NaHCO₃ solution (sat.), 1 wt %–HCl solution and NaCl solution (sat.) and dried over Na₂SO₄. After the gentle removal of the solvent using a rotary evaporator, the crude product was purified by column chromatography starting with pentane:EtOAc 3:1 and increasing to 0:1. After freeze-drying a yellowish, very viscous liquid was obtained as the final product (76%). Due to the highly reactive nature of CCMA all used solutions

and solvents were kept cool and BHT was added to inhibit polymerization during purification. The final product was stored in a freezer.

MS: calculated: $186.16 \text{ g mol}^{-1}$, found: 209.0425 [M + Na⁺], 395.0950 [2M + Na⁺].

¹H NMR (400 MHz, CDCl₃) [ppm] δ = 6.17 (s, 1H, C=CH₂), 5.68 (s, 1H, C=CH₂), 5.01 (m, 1H, CH), 4.61 (t, 1H, CH₂ cyclic carbonate), 4.44 (m, 1H, CH₂ cyclic carbonate), 4.36 (m, 2H, CH₂–O), 1.97 (s, 3H, CH₃); ¹³C NMR (101 MHz, CDCl₃) [ppm] δ = 166.62 (C=O methacrylate), 154.38 (C=O cyclic carbonate), 135.12 (C=CH₂), 127.29 (CH₂=C), 73.79 (CH cyclic carbonate), 66.05 (CH₂–O), 63.42 (CH₂ cyclic carbonate), 18.14 (CH₃). ATR-IR [cm^{−1}]: 2959 (s, CH₃, CH₂), 1792 (w, C=O cyclic carbonate), 1718 (m, C=O), 1640 (m, C=C), 1296 (w, ester), 1060 (w, ester), 811 (m, C=CH₂), 712 (m, C=CH₂).

2.3. Polymer synthesis

The polymers were prepared by mixing CCMA and 0.2 wt% BNP for the CCMA-homopolymer and OEGMA and CCMA in different molar ratios and 0.2 wt% BNP for the co-polymers, respectively. The well-stirred mixtures were then drawn into a film by coating a certain amount between two quartz glass plates using Mylar® foil type A (DU PONT FILMS) and a spacer to obtain a constant thickness of about 300 μm . The polymerization was performed using UV-light (UVACUBE 100, Hoenle) for one hour. After polymerization the films were dried in a glass oven (BÜCHI Labortechnik) under reduced pressure at 120 °C over night.

2.4. Gel polymer electrolyte preparation

Gel polymer electrolytes were prepared by gelling polymer disks with diameters of 7–10 mm with appropriate amounts of liquid electrolyte (1 M LiPF₆ in EC:DMC 1:1 w:w, LP30, Merck, battery grade, conductivity: 10.7 mS cm^{-1} [7]) to obtain a polymer:liquid electrolyte ratio of 1:5.5 corresponding to a composition of 15 wt% polymer and 85 wt% liquid electrolyte. To ensure complete soaking all disks were allowed to gel in sealed glass vessels for at least 24 h.

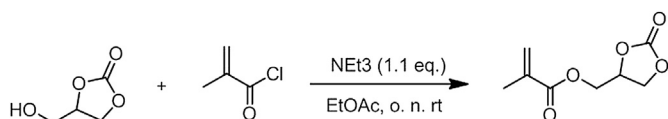
2.5. Thermal characterization

For Thermogravimetric Analysis (TGA) of the synthesized polymers and gel polymer electrolyte, a TGA Q5000 (TA Instruments) was used. Each sample was heated from room temperature to 600 °C with a heating rate of 5 K min^{-1} .

DSC measurements were carried out using a DSC Q2000 (TA Instruments). 10 mg of the polymer was put in a closed aluminum can. The sample was cooled from room temperature to -150 °C followed by an increase in temperature up to 120 °C with a heating rate of 10 K min^{-1} in a nitrogen atmosphere. Three cooling and heating cycles were conducted for every sample.

2.6. Electrochemical characterization

The conductivity of the gel polymer electrolytes was investigated in a sealed lab-made cell employing two stainless steel electrodes with a distance of 600 μm , which was assembled in an argon filled glove-box (MBraun, O₂ and H₂O content lower than 1 ppm). The temperature dependent conductivity of the gel polymer electrolyte between 5 °C and 55 °C was measured using a Solartron 1260 impedance analyzer, which was connected to a Solartron 1287A potentiostat/galvanostat and an environmental simulation chamber (MK 53, Binder GmbH). All measurements were performed in a frequency range of 10 Hz to 10 MHz with a voltage amplitude of 5 mV. The given conductivities are an average of three measurements.



Scheme 1. Synthesis of the monomer cyclic carbonate methacrylate (CCMA).

For the measurement of the oxidative stability the gel polymer electrolyte ($d = 12$ mm) was placed between a lithium counter ($d = 12$ mm) and a platinum working ($d = 12$ mm) electrode inside a Swagelok® cell with lithium as reference electrode. The oxidative stability was determined by linear sweep voltammetry using a Solartron 1287A potentiostat/galvanostat in a potential range from open circuit potential (OCP) to a maximum of 6.0 V with a scan rate of 0.1 V s^{-1} .

The rate capability and the cycling performance of the gel polymer electrolyte in lithium-ion batteries were evaluated using three-electrode cells (Swagelok®) with $\text{Li}(\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3})\text{O}_2$ (NCM, $d = 11$ mm) as cathode, graphite ($d = 12$ mm) as anode and lithium (Rockwood Lithium, 99.9%) as reference electrode. For these experiments the same electrodes like in Ref. [12] were used. The electrodes were pre-wetted with 20 μL of liquid electrolyte. For the determination of the rate capability of cells comprising the gel polymer electrolyte, charging was conducted with C/10 and the discharge rate was increased from C/10 to 5C in a potential range of 2.8–4.2 V vs. Li/Li^+ . The cycling performance of the gel polymer electrolyte was measured using a MACCOR series 4000 battery cyclers. The measuring protocol involved a resting step of the cell for 12 h, followed by charge and discharge at a rate of C/10 in the first three cycles to form a stable solid electrolyte interphase (SEI). Afterward, the cell was charged and discharged with a C-rate of C/2 in a voltage range of 2.8–4.2 V vs. Li/Li^+ .

For comparison all cell experiments were also conducted in cells comprising the liquid electrolyte (1 M LiPF_6 in EC:DMC 1:1) soaked in a Whatman separator (GF/D).

3. Results and discussion

3.1. Monomer and polymer synthesis and characterization

The synthesis of the monomer cyclic carbonate methacrylate (CCMA) ((2-Oxo-1,3-dioxolan-4-yl)methyl methacrylate) is shown in Scheme 1. By elimination of hydrochloric acid, glycerol 1,2-carbonate and methacryloyl chloride reacted to the desired product CCMA with a yield of 76% via an esterification. According to literature, the compound was identified via NMR and IR spectroscopy [14,15]. Additionally, the mass of the molecule was detected using mass spectrometry (MS).

The monomer CCMA was subsequently used for the synthesis of a homopolymer using UV-induced free radical polymerization. This polymerization method is the method of choice for the preparation of the polymer host as it is very fast and also cheap [4]. Additionally, the polymer is already obtained as homogeneous film after polymerization and therefore no additional film-forming step is needed. The glass transition temperature (T_g) of the resulting polymer film was determined via DSC measurements. The T_g of a polymer marks the transition from a rubbery into a glassy state. Therefore, the polymer is flexible above the T_g and hard and brittle below the T_g . The CCMA-homopolymer displayed a high T_g of 91°C (not shown here) resulting in brittle polymer films which were not

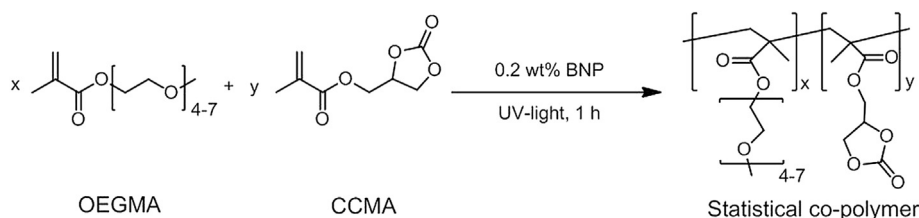
suitable for the preparation of gel polymer electrolytes. Therefore, statistical co-polymers comprising OEGMA as second monomer were synthesized. OEGMA was chosen as it provides the ability to decrease the T_g resulting in polymer matrices with increased flexibility. Various mixtures of CCMA, OEGMA and BNP as photoinitiator were prepared and polymerized via UV-induced free radical polymerization (see Scheme 2).

Polymer films with high amounts of CCMA were desirable as this component is expected to provide a good interaction with the liquid electrolyte due to its EC-related structure, thus, retaining it inside the matrix. Additionally, due to the high T_g of the homopolymer, it should be able to improve the mechanical properties of the resulting gel polymer electrolyte to allow its simultaneous use as electrolyte and separator. However, if the CCMA content is too high brittle polymer films are obtained whereas a too high amount of OEGMA leads to sticky polymer films with low mechanical stability. In order to optimize the properties of the statistical copolymers, polymers with molar ratios of OEGMA:CCMA of 2:1, 1:1, 1:2 and 1:1.2 were prepared and the T_g s of the resulting films were determined with DSC measurements. In Fig. 1 the results of the DSC measurements are shown. A molar ratio of 1:1.2 (OEGMA:CCMA) was found to be the best composition resulting in flexible and mechanically stable polymer films with a high amount of CCMA and a T_g of 5°C . Therefore, for all further experiments the statistical co-polymer OEGMA:CCMA 1:1.2 mol was used. A picture of the polymer film is shown in Fig. 2A.

The thermal stability of the statistical co-polymer was determined by TGA measurements, where the sample weight is monitored in dependency of the applied temperature. Thermal decomposition of the polymer starts at approximately 200°C (see Fig. 3). This is far above the temperature range in which lithium-ion batteries are commonly used. Therefore, it can be assumed that the polymer matrix is not limiting the applicability of the resulting gel polymer electrolyte at elevated temperature.

3.2. Gel polymer electrolyte

To determine the maximum amount of liquid electrolyte that can be immobilized in the polymer matrix gelling experiments were conducted by exposing the polymer films to an excess of liquid electrolyte. It was found that the prepared membranes are able to absorb and retain extraordinary amounts of liquid electrolyte. The statistical copolymer based on OEGMA and CCMA (1:1.2 mol) is able to immobilize 5.5 equivalents of liquid electrolyte based on its own weight resulting in gel polymer electrolytes with a composition of 15 wt% polymer matrix and 85 wt% liquid electrolyte. If the amount of liquid electrolyte is further increased partial leakage of the liquid electrolyte cannot be excluded. By the presence of the cyclic carbonate moiety in the polymer matrix, considerably improved gelling behavior can be achieved compared to other ethylene glycol-based polymer systems described in literature [12,16,17]. Even though the polymer membrane was able to absorb high amounts of liquid electrolyte, the gelled polymers



Scheme 2. Polymerization of OEGMA and CCMA by UV-induced free radical polymerization.

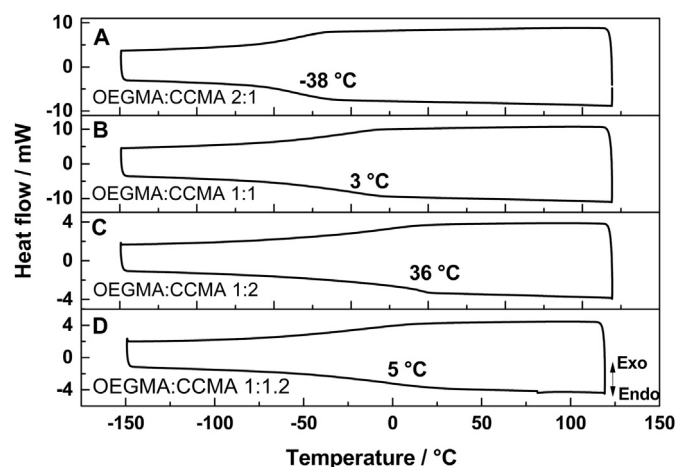


Fig. 1. DSC measurements of statistical co-polymers with different molar ratios of OEGMA:CCMA. Second cooling/heating cycles are shown. Cooling/heating rate: 10 K min⁻¹.

were obtained as self-standing, mechanically stable and flexible films, which can be applied as gel polymer electrolytes and separators in lithium-ion batteries (see Fig. 2B).

TGA experiments were also used for the investigation of the thermal stability of the prepared gel polymer electrolyte (Fig. 3). As mentioned before the ungelled polymer membrane starts to decompose at approximately 200 °C. In case of the gel polymer electrolytes not only the decomposition of the components but also the evaporation of the solvents of the liquid electrolyte is possible. For the liquid electrolyte a weight loss is already visible starting from 30 °C, due to the presence of the highly volatile linear carbonate DMC. Also in the TGA curve of the gel polymer electrolyte a weight loss starting at 30 °C is observable. Though, it seems that the kinetics of evaporation is slowed down due to the interaction between the polymer host and the solvent of the liquid electrolyte.

3.3. Electrochemical characterization

Another important parameter for electrolytes for lithium-ion batteries is a high ionic conductivity. In case of gel polymer electrolytes the conductive properties mainly depend on the amount of liquid electrolyte that can be absorbed in the polymer matrix.

The temperature dependent conductivity of the gel polymer electrolyte was determined in a temperature range between 5 °C and 55 °C (see Fig. 4). With increasing temperature the conductivity

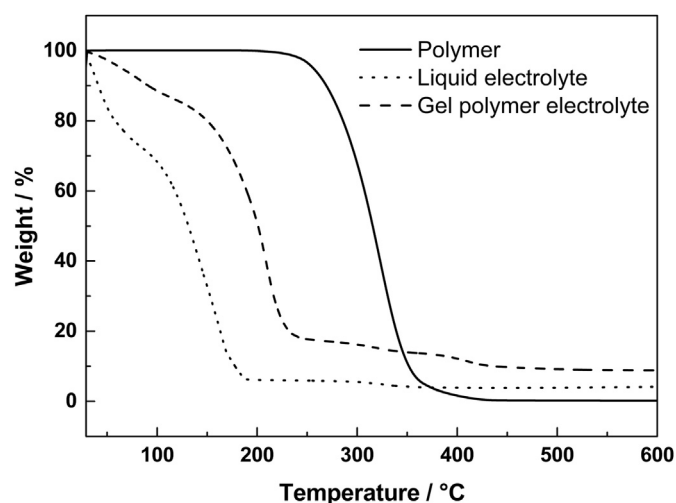


Fig. 3. TGA measurements of the polymer membrane (OEGMA:CCMA 1:1.2 mol), the liquid electrolyte 1 M LiPF₆ in EC:DMC 1:1 w:w and the gel polymer electrolyte (15 wt% polymer + 85 wt% liquid electrolyte). Heating rate: 5 K min⁻¹.

increases up to 5.0 mS cm⁻¹ at 55 °C. Furthermore, even at a low temperature of 5 °C still a conductivity of 1.2 mS cm⁻¹ is obtained. As the polymer membrane is able to host large amounts of the liquid electrolyte a high ionic conductivity of 2.3 mS cm⁻¹ at 25 °C can be achieved, which is comparable or even higher than the conductivities reported for other gel polymer electrolyte systems [16,18–21]. Reiche et al. studied an ethylene glycol–dimethacrylate-based gel polymer electrolyte system, which reached an ionic conductivity of around 10⁻¹ mS cm⁻¹ at 25 °C [18].

As the conductivity of the gel polymer electrolyte is in the mS cm⁻¹ range like that of liquid electrolytes it is assumed that it is high enough for use in lithium-ion batteries.

The oxidative stability of the gel polymer electrolyte, which is of special interest as polymers comprising oligo ethylene glycol side chains were reported to decompose at higher potentials [22], was determined using linear sweep voltammetry. In Fig. 5 the oxidative stability of the gel polymer electrolyte in comparison to that of the liquid electrolyte is shown. Both electrolytes show a similar

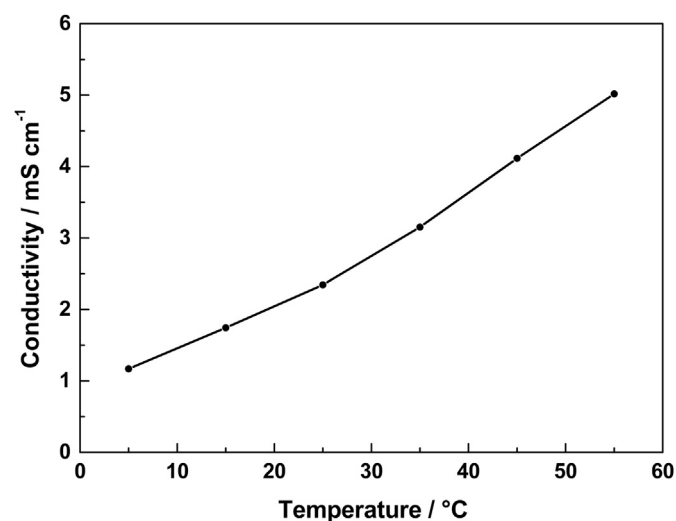


Fig. 4. Temperature dependent conductivity of the gel polymer electrolyte (15 wt% polymer (OEGMA:CCMA 1:1.2 mol) + 85 wt% liquid electrolyte (1 M LiPF₆ in EC:DMC 1:1 w:w)) in the range of 5–55 °C.

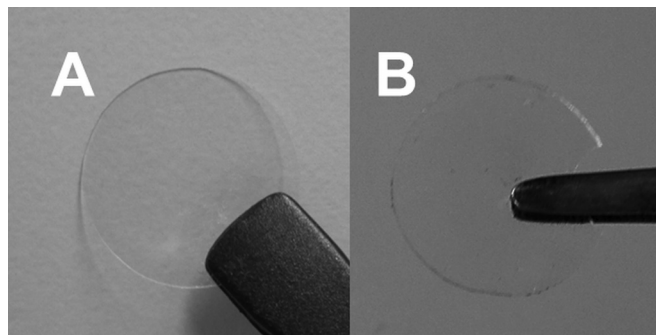


Fig. 2. Photographs of the statistical co-polymer (OEGMA:CCMA 1:1.2 mol) before (A) and after (B) swelling in 5.5 equivalents of liquid electrolyte (1 M LiPF₆ in EC:DMC 1:1 w:w) resulting in gel polymer electrolytes with the composition 15 wt% polymer + 85 wt% liquid electrolyte.

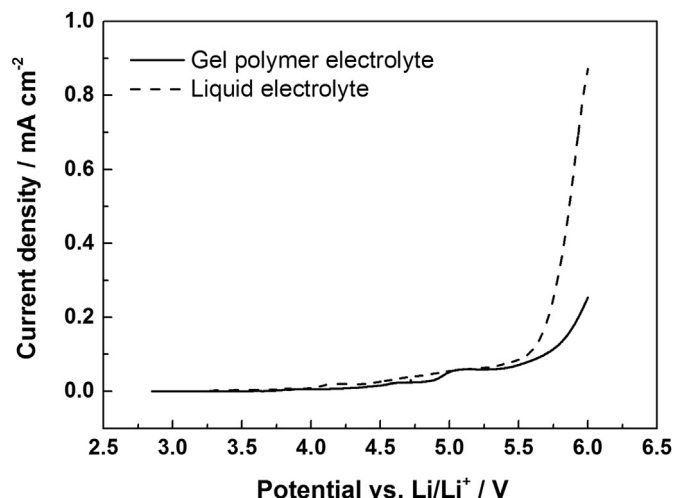


Fig. 5. Oxidative stability of the gel polymer electrolyte (15 wt% polymer (OEGMA:CCMA 1:1.2 mol) + 85 wt% liquid electrolyte (1 M LiPF₆ in EC:DMC 1:1 w:w)) and the liquid electrolyte. Working electrode: Pt. Reference and counter electrode: Li. Scan rate: 1 mV s⁻¹.

behavior. An increase in current density is observed for both electrolytes in the same potential region. The results indicate that the investigated gel polymer electrolyte displays an oxidative stability, which is comparable to that of the liquid electrolyte that is immobilized in the gel.

To determine the rate capability of the gel polymer electrolyte, a C-rate experiment was carried out. In the first three cycles the cells were charged and discharged with C/10 to allow the formation of a stable SEI. Afterward, charge was performed at a C-rate of C/10 and discharge was conducted with varying rates from C/10 to 5C. As comparison, the same measurement was conducted with cells comprising the liquid electrolyte in combination with a *Whatman* separator. In Fig. 6 the results for both electrolytes are shown. At lower C-rates of C/10, C/5 and C/2 the cells containing the gel polymer electrolyte display capacities of 138 mAh g⁻¹, 134 mAh g⁻¹ and 126 mAh g⁻¹, respectively. Also at a moderate high C-rate of 1C the capacity is still as high as 114 mAh g⁻¹. Just at higher C-rates of 3C and 5C both electrolytes display reduced performance. This is particularly true for the cells comprising the gel polymer electrolyte, which reach 45 mAh g⁻¹ and 31 mAh g⁻¹ at 3C and 5C, respectively. This can be explained by its lower ionic conductivity compared to that of the liquid electrolyte.

Constant current cycling experiments were carried out to determine the long-term cycling stability of the gel polymer

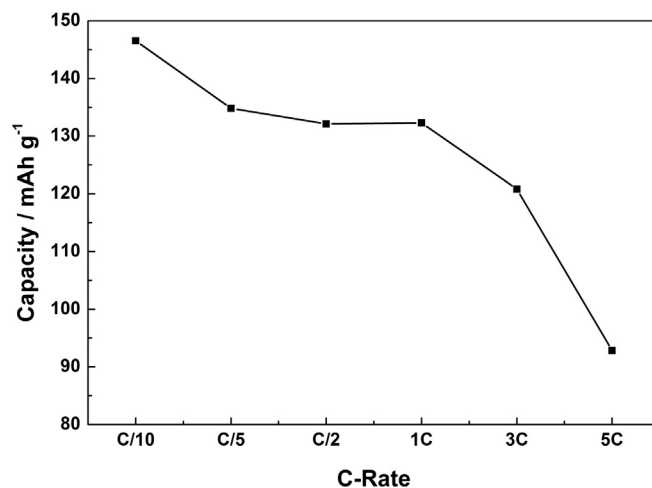
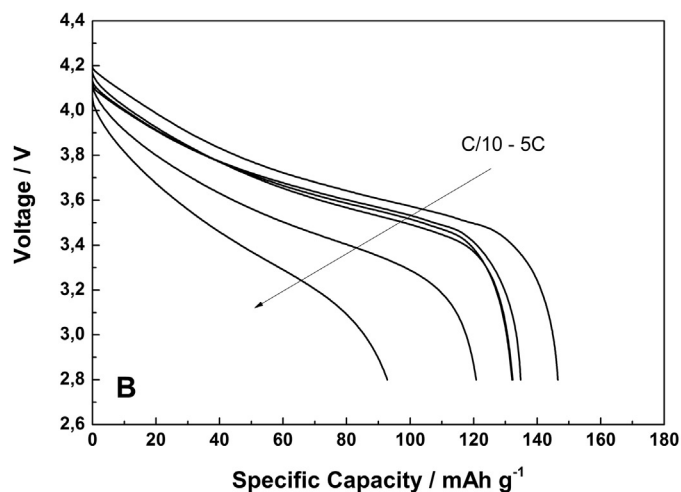
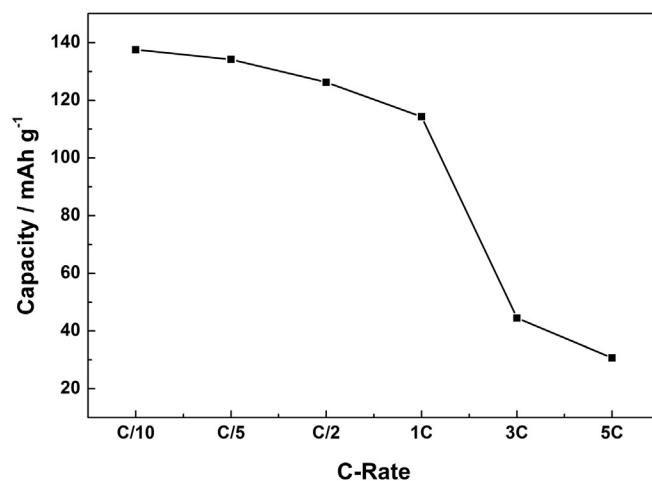
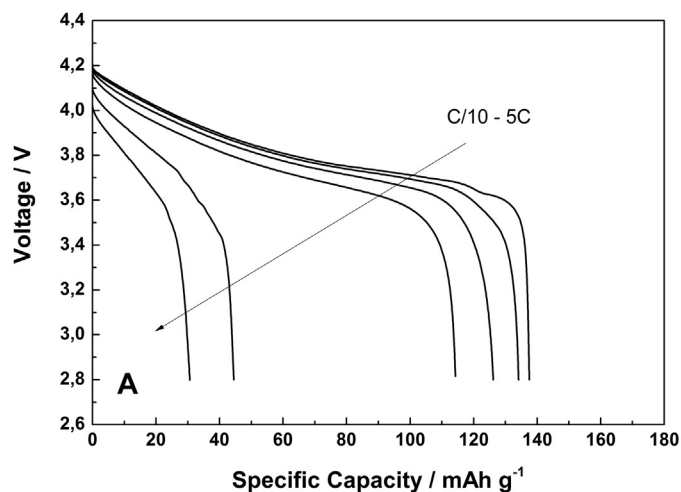


Fig. 6. Evaluation of the rate capability of the gel polymer electrolyte (15 wt% polymer (OEGMA:CCMA 1:1.2 mol) + 85 wt% liquid electrolyte (1 M LiPF₆ in EC:DMC 1:1 w:w)) (A) and the liquid electrolyte (B) in NCM/graphite full cells. Charge: C/10, discharge with varying rates (C/10–5C). Potential range 2.8–4.2 V vs. Li/Li⁺.

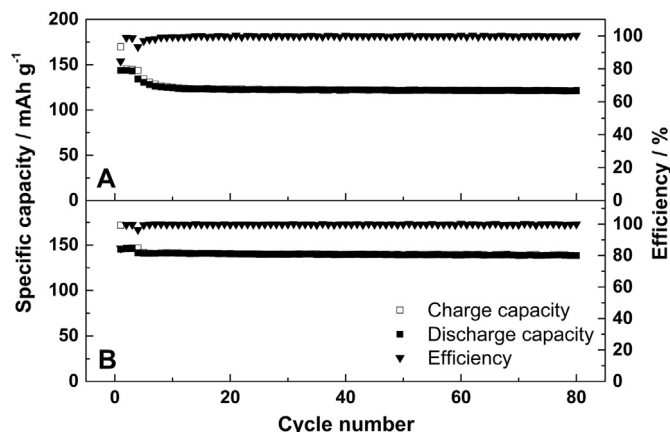


Fig. 7. Constant current cycling of NCM/graphite full cells comprising the gel polymer electrolyte (15 wt% polymer (OEGMA:CCMA 1:1.2 mol) + 85 wt% liquid electrolyte (1 M LiPF₆ in EC:DMC 1:1 w:w)) (A) and the liquid electrolyte (B). Potential range: 2.8–4.2 V vs. Li/Li⁺.

electrolyte. Therefore, cells with NCM as cathode and graphite as anode material with a lithium reference electrode were cycled in a potential range of 2.8–4.2 V vs. Li/Li⁺. In the first three cycles the cells were charged and discharged at C/10 to form a stable SEI resulting in efficiencies of 85%, 99% and 99% (see Fig. 7A). Afterward the charge and discharge was carried out at a C-rate of C/2. Starting from the fourth cycle a reduced capacity is observed which is caused by the C-rate increase from C/10 to C/2. In the following cycles the cells show a very stable cycling behavior over 80 cycles with efficiencies of >99.5%. After 80 cycles the cells still reach a capacity of 122 mAh g⁻¹ corresponding to a capacity retention of >91%. For comparison also cells with the liquid electrolyte were tested under the same conditions (see Fig. 7B). Similar capacity retentions were obtained. The cells containing the liquid electrolyte display slightly higher capacities compared to that comprising the gel polymer electrolyte, which is due to the lower ionic conductivity of the gel polymer electrolyte compared to the liquid electrolyte. The results of the constant current cycling experiments indicate that the investigated gel polymer electrolyte can be used with standard materials in lithium-ion batteries.

4. Conclusion

In this study we reported on the synthesis of a new polymer host material for gel polymer electrolytes for lithium-ion batteries. The statistical co-polymer OEGMA:CCMA (1:1.2 mol) was polymerized by UV-induced free radical polymerization and the polymer was obtained as self-standing, mechanically stable and flexible

film, which allowed the use of the polymer as host material in gel polymer electrolytes. Due to the huge amount of liquid electrolyte (1 M LiPF₆ in EC:DMC 1:1 w:w) that can be immobilized and retained in the polymer host structure (550% of its own weight) a high ionic conductivity of 2.3 mS cm⁻¹ at 25 °C was achieved. Additionally, the gel polymer electrolyte showed a good C-rate capability at moderate high C-rates and constant cycling performances in NCM/graphite full cells, comparable to that displayed by the standard liquid electrolyte.

Considering these results, the investigated gel polymer electrolyte appears as a promising candidate for use in lithium-ion batteries.

Acknowledgments

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